REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

1. AGENCY USE ONLY *(Leave Blank)*	2. REPORT DATE	3. REPORT TYPE AND DATES COVERED
	30 September 98	Progress Report: 1 July 98 - 30 September 98

4. TITLE AND SUBTITLE
Processing of Nanocrystalline Nitrides and Oxide Composites

5. FUNDING NUMBERS
G - N00014-95-1-0626

6. AUTHORS
Jackie Y. Ying
Martin L. Panchula

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
Department of Chemical Engineering
Massachusetts Institute of Technology
77 Massachusetts Avenue, Room 66-544
Cambridge, MA 02139-4307

8. PERFORMING ORGANIZATION REPORT NUMBER

9. SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES)
Office of Naval Research
800 North Quincy Street
Ballston Tower One
Arlington, VA 22217-5660

10. SPONSORING / MONITORING AGENCY REPORT NUMBER

11. SUPPLEMENTARY NOTES

12a. DISTRIBUTION / AVAILABILITY STATEMENT

Approved for public release; distribution unlimited

12b. DISTRIBUTION CODE

13. ABSTRACT *(Maximum 200 words)*

We have examined various synthesis parameters for producing nanocrystalline aluminum nitride and established maps relating surface area and crystallite size to synthesis conditions. Preliminary analysis of the powder compacts and initial sintering studies show a great deal of potential. Green compacts with a very fine average pore size of 5.5 nm were obtained readily. Densities of 96% were achieved without sintering additives after 30 minutes of pressureless sintering at 1900°C in flowing nitrogen. We are currently examining sintering behavior at lower temperatures in detail. Future work will also involve chemical and thermal characterization of the sintered ceramics.

14. SUBJECT TERMS
Nanocrystalline, Aluminum Nitride, Synthesis, Sintering

15. NUMBER OF PAGES
4

16. PRICE CODE

17. SECURITY CLASSIFICATION OF REPORT	18. SECURITY CLASSIFICATION OF THIS PAGE	19. SECURITY CLASSIFICATION OF ABSTRACT	20. LIMITATION OF ABSTRACT
UNCLASSIFIED	UNCLASSIFIED	UNCLASSIFIED	UL

"Processing of Nanocrystalline Nitrides and Oxide Composites"

19981014 041

Technical Report on ONR Grant No. N00014-95-1-0626
for the period of July 1, 1998 - September 30, 1998

Jackie Y. Ying
Department of Chemical Engineering
Massachusetts Institute of Technology
Room 66-544, 77 Massachusetts Avenue
Cambridge, MA 02139-4307
Tel: (617) 253-2899
Fax: (617) 258-5766

Nanocrystalline Aluminum Nitride

Nanocrystalline aluminum nitride has been successfully synthesized in the forced flow reactor through the use of ammonia as the nitriding gas [1]. Recent efforts have focused on the preparation of larger amounts of material and improving the quality of the aluminum nitride produced. Both of these endeavors involved some modifications to the reactor design, such as (1) addition of baffles in the hot zone to produce a more laminar flow, (2) introduction of an argon purge into the crucible to prevent nitridation of the melt, and (3) addition of a nitrogen line downstream from the filter assembly so the filter can be backflushed. With these changes, the yield was significantly improved, and the evaporation experiments can run continuously until the aluminum supply is exhausted.

Studies relating the reactor operating conditions and AlN powder properties have also been performed, although with the most recent changes made to the reactor the specific results may be affected. As observed by other researchers using similar low-pressure reactors, the velocity and overall pressure can have a large effect on the degree of agglomeration, crystallite size, and surface area. Generally, it has been found that high quench rates and low reactor pressures produce less agglomerated and smaller particles. Our research results support these observations especially with regard to the pressure - crystallite size relationship shown in Figure 1. The surface area data in Figure 1 appear scattered, however this is because in addition to the pressure variation shown on the ordinate, the volume of gas, and therefore the velocity, are changed as well. This may be more clearly seen in Figure 2 where the surface area is shown as a function of gas velocity and reactor pressure. In our reactor, it appears that there is a maxima in the surface area - gas velocity relationship that shifts depending on the overall reactor pressure. Crystallite size appears to be primarily a function of reactor pressure and is relatively unaffected by gas velocity, which makes sense considering that the clusters are formed directly over the melt and rapidly react to form stable AlN crystallites before being swept into the gas stream.

Aluminum Nitride Processing

The air-free handling of nanocrystalline AlN is another important step in producing the desired dense, pure nitride ceramics. We have performed only preliminary sintering studies thus far; the oxygen content of these samples has yet to be evaluated along with the optimization of the processing parameters. The pore size distribution of a sample before and after compaction is shown in Figure 3. The narrow pore size distribution of the cold isostatically pressed (CIPed) sample suggests that the powder is of high quality with very few large agglomerates and should sinter very well. In fact, a sample of nanocrystalline AlN pressed at 62 MPa, CIPed at 410 MPa, and pressurelessly sintered without sintering aids under a flowing nitrogen stream at 1900°C achieved 96% of theoretical density after 30 minutes. This is quite a promising result since even with oxide additives and liquid-phase sintering, AlN is normally sintered at temperatures >1800°C to achieve high density and good thermal conductivity.

Future Work

Future efforts will be focused on the processing and sintering of nanocrystalline AlN to produce ceramics with low oxygen content and high thermal conductivity. The oxygen, nitrogen, and aluminum content of the powder and dense specimens will be determined through a combination of fast neutron activation analysis (FNAA) and inductively coupled plasma - atomic emission spectroscopy (ICP-AES). These methods will allow direct air-free analysis of the sample. The thermal conductivity experiments will be performed on the new laser flash thermal diffusivity equipment at the Naval Research Laboratory.

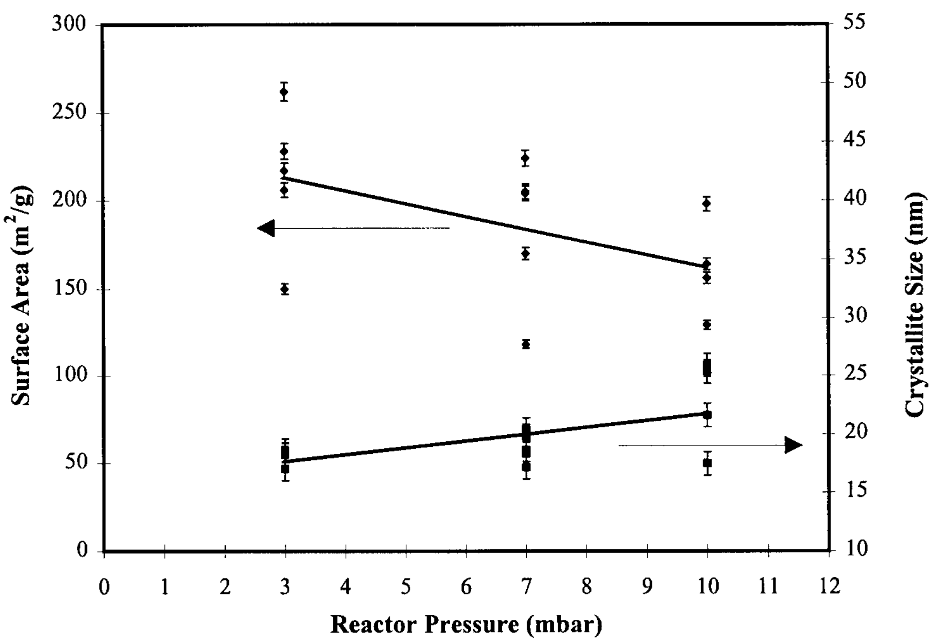

Figure 1. Surface area and crystallite size as a function of reactor pressure.

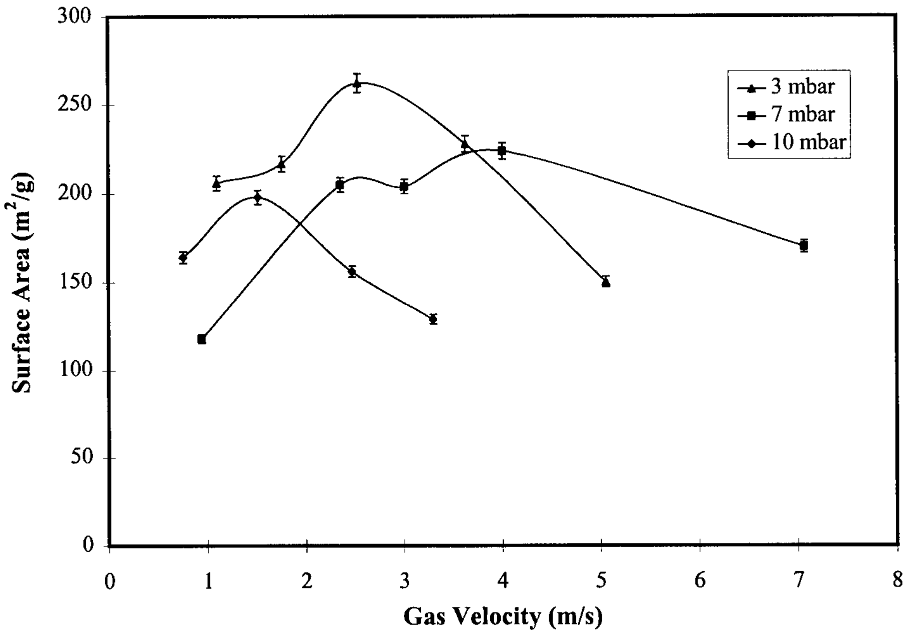

Figure 2. Surface area as a function of gas velocity and reactor pressure.

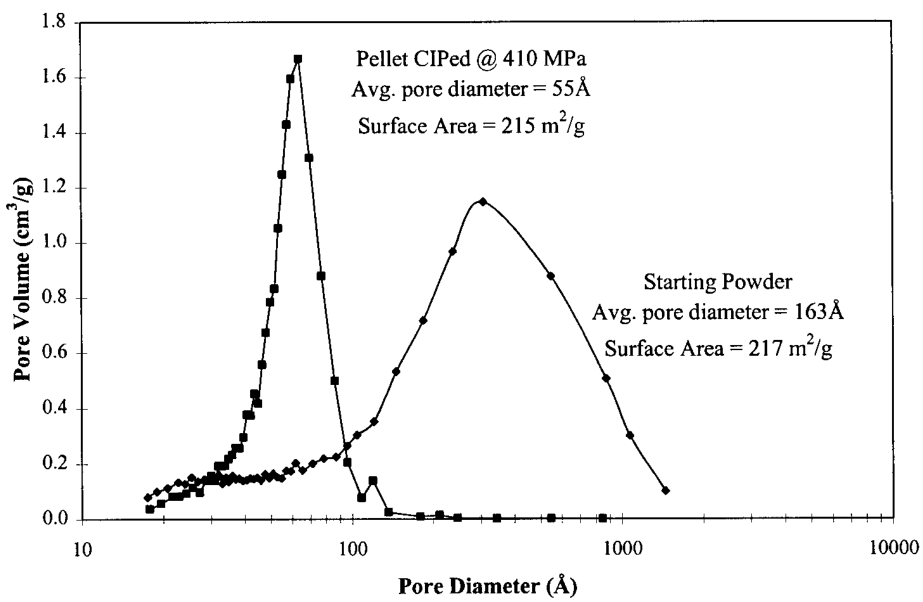

Figure 3. Pore size distribution (dV/dlogD) of loose powder and unsintered compact.

---

[1] Panchula, M.L. and Ying, J.Y., M.I.T., unpublished results.